Validity of effective medium theory in multilayered hyperbolic materials

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Metal-dielectric multilayers can be designed to exhibit remarkable optical properties, including negative refraction for subwavelength superlensing. In this study, the applicability of the medium-homogenized effective medium theory (EMT) in place of multilayered thin-film optics is examined. Three metal-dielectric material and thin film thickness combinations that give rise to hyperbolic dispersion in different spectral regions are considered. In addition to investigating the radiative properties, the energy streamline method is used to determine the refraction angle and lateral displacement of rays. The electromagnetic fields inside the films are depicted to illustrate the coherent effect or the lack thereof. The radiative penetration depth is profiled to understand the effectiveness and limits of such multilayers in optical manipulation. The conditions and mechanism for the breakdown of EMT are elucidated in this case study.

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1. Introduction

One-dimensional periodically stratified media, or multilayers, are often used for antireflection and wavelength-selective thermal radiation applications [1,2]. Recently, negative refraction has been demonstrated using carefully designed metal-dielectric or semiconductor multilayers due to their effective anisotropic permittivities [3–5]. It is easier to construct nonmagnetic hyperbolic metamaterials of single negative electrical permittivity, as the free electron motion is confined in one spatial direction [5]. This anisotropy of the combined homogeneous material slab is accomplished by patterning or depositing isotropic constituent layers in alternating fashion. Hyperbolic metamaterials are developed and refined to achieve sub-diffraction imaging through subwavelength nanostructures or nanofilms [6–8]. Other thermal engineering applications of hyperbolic multilayers are coherent radiative emission, electromagnetic waveguiding, and near-field thermophotovoltaic or thermal rectification [9–14].

In this study, the energy streamlines and radiative properties are closely inspected for various experimentally achieved multilayers to reevaluate and validate existing modeling assumptions. A metal-dielectric multilayer in the visible wavelengths and two semiconductor multilayers in the infrared are considered. Semiconductor multilayers consist of alternating undoped and metal-alloy layers, the latter of which the doping concentration can be tuned for desirable spectral response. The radiative behaviors are studied using the transfer matrix method (TMM) and effective medium theory (EMT). TMM is formulated using a system of linear equations for each constituent layer, with given isotropic permittivity (dielectric function) and thickness. On the other hand, EMT treats the stratified layered structure as a homogeneous medium with an effective permittivity tensor. This requires that the thicknesses of the constituent layers be much smaller than the wavelength of consideration. This work examines the radiative properties, energy streamlines, and electromagnetic fields of varying multilayer configurations and their spectral ranges. The outcomes should uncover conditions when EMT is not suitable for estimating multilayer properties, and describe the mechanisms or quantitative criteria for the breakdown of EMT in specific applications.

2. Methods

Fig. 1 illustrates the multilayers containing alternating dielectric (d) and metal (m) media. The period Λ is the sum of one of dielectric (d a) and metal (d m) layers’ thicknesses. The incidence angle from vacuum of unity permittivity is denoted as θ i. After N multilayer periods, the medium of outgoing rays is also a vacuum. The transfer matrix method (TMM) has been used to determine the radiative properties and spatially-varying Poynting vector in layered thin films [1,2]. In the present study, only transverse magnetic (TM) waves that may support negative refraction are considered. The Fresnel coefficients for a uniaxial medium can be obtained from Ref. [15] to modify the TMM so that each layer can be a uniaxial medium. The consecutive products of the matrix set determine the field amplitudes inside each layer by applying boundary conditions. Analytical expressions for the field amplitudes with 3 layers, the center containing a thin film, can be found in Refs. [16,17].
The known electric and magnetic fields are determined by applying Maxwell’s equations to obtain the time-averaged Poynting vector, whose components are determined by

\[ S_x = 0.5Re(E_x \times H_y^*) \] (1)

and

\[ S_z = 0.5Re(E_z \times H_y^*) \] (2)

where * denotes the Hermitian complex transform of the function. The Poynting vector is only a function of z, since the x and temporal terms are cancelled. A simple spatial extrapolation method of the Poynting vector maps the energy streamlines (ESL) [16]. The planes perpendicular to the energy streamlines represent the domain of phase fronts. The radiative properties, reflectance and transmittance, are determined by the ratios of field amplitudes.

A way to homogenize a medium into a single slab is to use EMT. For periodic multilayered structures, Rytov [18] developed analytical formulation to approximate the stratified medium with a uniaxial dielectric tensor under the condition when the wavelength of the incidence electromagnetic radiation is much longer than the unit cell, in this case defined as either \( d_d \) or \( d_m \). The above expressions are suitable for anisotropic metal and dielectric layers as well, forming a class of hyperbolic metamaterials [19–21]. The applicability of EMT for near-field thermal radiation was discussed by Liu et al. [14]. The anisotropic permittivity of the multilayer is given in the ordinary direction (perpendicular to the optical axis along z) and extraordinary direction (parallel to optical axis), viz. [13,20],

\[ \varepsilon_O = f \varepsilon_{m,0} + (1-f)\varepsilon_{d,0} \] (3)

and

\[ \varepsilon_E = \frac{\varepsilon_{m,\infty} \varepsilon_{d,\infty}}{f \varepsilon_{d,\infty} + (1-f)\varepsilon_{m,\infty}} \] (4)

Here, the filling ratio is the quotient of the slab thickness of metal to period size \( f = d_d/A \). Typically, the filling ratio is bounded between 0.2 and 0.8, else the radiative properties between TMM and EMT have found to diverge [20]. Since EMT is only a function of the relative constituent layer thicknesses, the period can be successively varied in the calculation to examine the validity of EMT by comparing with the calculations based on TMM.

Since metals usually possess negative permittivity, the overall permittivity of the multilayer given by Eqs. (3) and (4) may also be negative. The two equations allow two types of electrically hyperbolic dispersions: Type I and Type II. Type I is defined by \( \varepsilon_O > 0 \) and \( \varepsilon_E < 0 \) [5,19,20]. Type II is defined by \( \varepsilon_O < 0 \) and \( \varepsilon_E > 0 \). Only the anomalous dispersion in Type I hyperbolic metamaterials enable negative angle refraction of propagating waves (solely real wavevectors) inside the homogenized medium [21–23].

3. Results and Discussion

Alternating metal and dielectric layers have achieved left-handed response in visible wavelengths, which benefits toward optical waveguiding and subdiffraction imaging [5,8,11]. These simply-constructed thin films typically contain evaporated nanometers-thick metal layers over large surface areas, consisting of elements such as gold or silver [24,25]. The dielectric layers within the course of wavelengths of interest are resonance-free, thus good impedance matching with air and relatively low radiative attenuation [11]. On the other hand, instead of metal layers, which may contain enormous loss due to electron carrier absorption, semiconductor layers may be substituted in. Semiconductors are simple to deposit, and once formed, can have adjustable electron density or doping concentrations to give more or less metallic behavior [26,27]. Examples of dopable semiconductor materials are metal nitrides, silicon, germanium, indium, and many others [28,29].

3.1. Choice of materials

To elucidate the differences of multilayer compositions toward the radiative properties, three multilayers with known material properties are compared. Three metal- or semiconductor-dielectric multilayers are presented, with their dielectric function and Drude model parameters listed in Table 1. The first material system is a hyperbolic multilayer containing silver (Ag) and rutile (TiO2), which was reported in Ref. [25] to demonstrate negative refraction in the UV to visible wavelengths. The fabricated structure consisted of three MDMDM units, with each layer having approximately \( d_{d,m} = 30 \) nm in thickness. In the simplifying case presented here, the alternating unit is just DM. Equivalently, \( N = 8 \) according to the multilayer geometry shown in Fig. 1. Semiconductor-dielectric layers were employed in Ref. [26] in which the former material is achieved by doping zinc oxide with aluminum (AZO-ZnO). The fabricated thin film was reported to have 16 alternating layers \( N = 8 \), each 60 nm thick. In this particular study, the doping concentration of the AZO layers is chosen to be \( 4.3 \times 10^{20} \text{ cm}^{-3} \). The semiconductor substitute for metal offers tunability by means of doping, and has less loss while offering negative permittivity. Furthermore, fabrication of doped semiconductors is more integrated, and does not require very thin layer deposition. For this configuration, the semi-continuous boundary between layers is delineated and controlled by the diffusion process of dopants. Another type of semiconductor-dielectric multilayers was proposed in Ref. [27], which consist of aluminum or gallium-doped indium arsenide (Al0.48In0.52As-In0.53Ga0.47As). The

![Fig. 1. Illustration of a periodic metal-dielectric multilayer. The incident and outgoing media are vacuum.](image_url)
doping concentration for most cases was set to $7.5 \times 10^{18} \text{ cm}^{-3}$. Here, the layers are 80 nm thick, and all alternating layers added to a 8.1 μm-thick slab ($N \approx 50$). For such relatively thick films, the radiative properties can be obtained by ray tracing method of multiply-reflected specular reflectance coefficients [2]. This assumes the incidence wavelength is comparable to or smaller than the film thickness, and approaches the incoherent limit where interference effects are neglected.

3.2. Dielectric functions

Fig. 2 plots the ordinary and extraordinary components of the dielectric function of the three multilayers. The wavelength regions at which the multilayers transition from Type I to Type II hyperbolic dispersion is distinct. The Type I hyperbolic regime is highlighted in blue, and Type II in yellow. The dielectric functions determined by EMT all have filling ratios of $f = 0.5$. In this case, $|\varepsilon_m^o| < |\varepsilon_m^e|$ must be true for Type I, and $|\varepsilon_m^o| > |\varepsilon_m^e|$ must

![Image of Fig. 2: Dielectric functions of the metal- or semiconductor-dielectric multilayers: (a) and (b) Silver-Rutile (Ag-TiO$_2$) real and imaginary components, respectively; (c) and (d) Aluminum-doped Zinc Oxide (AZO-ZnO); (e) and (f) Aluminum or Gallium Indium Arsenide (AlInAs-InGaAs). The middle (blue online) shaded region corresponds to $\varepsilon_m^o > 0$ and $\varepsilon_m^e < 0$, and the right (yellow online) shaded region corresponds to $\varepsilon_m^o < 0$ and $\varepsilon_m^e > 0$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)](image-url)
be true for Type II, when considering the constituent materials' permittivity magnitudes. The Ag-TiO$_2$ multilayers, shown in Fig. 2(a) and (b) makes the transition at $\lambda = 0.44 \mu m$. The magnitude of $\varepsilon'_E$ for this multilayer is remarkably greater than $\varepsilon'_O$. Near the transition between Type I and Type II, $\varepsilon'_E$ spikes due to the near-singularity of Eq. (4) as Eq. (3) approaches zero. The imaginary component, or loss in the ordinary direction is given by $\varepsilon''_O = 0.5\varepsilon''_m$. The parameter in the extraordinary component can be estimated to $\varepsilon''_E = 2\varepsilon''_O \varepsilon''_m$ at the peak where $|\varepsilon''_m| \approx |\varepsilon''_d|$. The dielectric function of the AZO-ZnO multilayers is shown in Fig. 2(c) and (d). The transition between Type I and II has a gap near $\lambda = 2.5 \mu m$, so that a narrowband region is double positive permittivity. Semiconductor multilayers have hyperbolic regimes in the mid- to far-infrared. Unlike the previous case with metal-dielectric multilayers, the hyperbolic regions hold relatively small $\varepsilon''_E$. The dielectric function of the indium arsenide multilayers is shown in Fig. 2(e) and (f). For this material combination, the Type I and II transition wavelength is in the

**Fig. 3.** Energy streamlines in their respective hyperbolic Type I and Type II wavelengths: Ag-TiO$_2$ multilayer thin films of $d = 480$ nm thickness at (a) $\lambda = 400$ nm, and (b) $\lambda = 600$ nm; AZO-ZnO multilayers of $d = 960$ nm thickness at (c) $\lambda = 2.0 \mu m$, and (d) $\lambda = 3.0 \mu m$; AlInAs-InGaAs multilayers of $d = 8 \mu m$ thickness (e) $\lambda = 9.5 \mu m$, and (f) $\lambda = 15 \mu m$. The incidence angle is $\theta_i = 40^\circ$. 

far-infrared, around $\lambda = 11 \mu m$. Since the dielectric component is relatively large, 10 as compared to 4.0, the imaginary component $\varepsilon''$ has a more pronounced peak.

3.3. Energy streamlines

The energy streamlines of the aforementioned multilayers are shown in Fig. 3. The TM wave ESLs all have incidence angles at $\theta_i = 40^\circ$, which correspond to the simulated beam field maps of Refs. [26] and [27]. The standard case for layer thickness is given alongside a case where the thickness of the layers is either increased or decreased. In Fig. 3(a), the standard case for Ag-TiO$_2$ multilayers is $d_{d,m} = 30$ nm. The wavelength used concerning Type I hyperbolic dispersion is $\lambda = 400$ nm, which is more than an order magnitude spatially larger than the thickness of constituent layers. However, the jagged ESL determined by TMM is remarkably different from that by EMT (dotted). Upon closer look, the dielectric layers swing the ESL toward the positive x direction, and the metal layers toward negative x. Closer to the exiting medium inside surface $(z/d = 1)$, the ESL becomes wavy within each layer. This signifies evanescent wave tunneling in the thin dielectric media between the near-field coupled plasmonic metal surfaces [14]. The layer thickness is then reduced to 12 nm, which gives ESL fitting better to that determined by EMT. The streamlines for the same multilayer thin film at a hyperbolic Type II wavelength are shown in Fig. 3(b), which show positive angle of refraction. As well, 30

![Fig. 4](attachment:image.png)

Fig. 4. Transmittance (solid lines and circles) and reflectance (dotted lines and open circles) profiles as functions of incidence angle. The wavelengths and thin film thicknesses correspond to those in Fig. 3.
nm-thick dielectric and metal layers contribute to stronger deviation from the streamline calculated by EMT. The standing waves generated in the incidence medium due to reflection cause the EMT-predicted streamlines to curl away.

The Type I and Type II regime ESL of AZO-ZnO multilayers are shown in Fig. 3(c) and (d), respectively. Despite having thicker layers and evident jaggedness, the ESLs hold very well to that estimated by EMT. Note the wavelength-to-constituent thickness ratios are within the same order as those presented in the previous case (i.e., $\lambda / d_{\text{m}} > 10$). In the case when $d_{\text{m}} = 120$ nm, only 4 periods are contained the thin film but yet maintains no obviously visible waviness or deviations. As well, at the Type II wavelength, no large shift from EMT is observed in the ESL determined by TMM. The closeness of these ESLs means oblique light beams traveling through multilayer films can be precisely spatially located, which is favorable toward development of demanding optical components. So far, semiconductor multilayers could mean better prediction by effective medium, which is likely further improved by the fabrication of a continuous medium with no roughness inclusions between layer interfaces. However, in Fig. 3(e), the ESL profiles for the indium arsenide multilayers show large deviations for very thick layers ($d_{\text{m}} = 500$ nm, $N = 8$). Despite having a comparable $\lambda / d_{\text{m}}$ ratio (17 versus 19) to that in the AZO-ZnO multilayers, EMT does not precisely predict the lateral shift in negative angle of refraction. Furthermore, the ESL for very thick layers in the Type II regime show a minute separation from that predicted by EMT. In fact, the $\lambda / d_{\text{m}}$ ratio in the Type II wavelengths between the AZO-ZnO and indium arsenide multilayers favors the latter (25 versus 30). In the case presented by the authors of Ref. [27], the large number of thin layers ($d_{\text{m}} = 80$ nm, $N = 50$) gives near-perfect matching with the ESL determined using EMT.

The transmittance and reflectance profiles in terms of the incidence angle, as shown in Fig. 4, also show significant differences between EMT and both cases calculated using TMM. The absorbance in the EMT homogenous medium accounts for most of the loss in transmittance. In Fig. 4, for example, transmittance is greatest and reflectance is near zero for EMT. The misleading lack of reflection is due to the good index matching ($\epsilon_{\text{i}} \approx 1.0$) at that particular wavelength [20]. On the other hand, transmittance is very poor in the wavelength selected for Type II, as seen in Fig. 4(b), where the transmittance is slightly favored toward the thicker layers. This is due to the reduced destructive interference between waves at fewer interfaces within the thin film medium. Considering the semiconductor multilayers, the transmittance profiles agree better between EMT and TMM of most layer thicknesses. Fig. 4(c) and (d) demonstrate both agreement between methods and modest transmittance or low reflectance in the AZO-ZnO multilayers. These characteristics are not all shared with the semiconductor multilayers described by Ref. [27], since significant differences between thin and thick layers are again evident in Fig. 4(e) and (f). Generally, radiative properties of multilayers containing very thick constituents wander quite a bit from that estimated by EMT.

3.4. Conditions for valid use of EMT

To better understand the deviations and good agreement of radiative properties in ZnO multilayers, some definitions and assumptions need to be established or revisited. As mentioned, the size of either metal or dielectric layer must be smaller than the characteristic wavelength. This is satisfied for all cases, where the smallest $\lambda / d_{\text{m}}$ ratio is 13.3. Agreement between EMT and TMM streamlines is certainly improved as this ratio is increased, but the limitations of deposition methods restrict nanometers-thick layers. The exact formulation for the thickness limit for the appropriate use of EMT is given by the Bloch wave theory [19]. The existence of propagating Bloch waves is given by the inequality, $k_d < k_{\text{z,EMT}}$, where $k_d$ and $k_{\text{z,EMT}}$ are defined in each homogeneous medium. The $\pi$ angle is dictated by the phase reversal of coherent radiation. The $z$-axis wavevector for an isotropic layer is determined by $k_z = \sqrt{k_d^2 - k_{\text{z,EMT}}^2}$. Since the permittivity of metal layer is largely negative, giving an almost purely imaginary component of $k_d$, only the results using dielectric layer are compared. For the Ag-TiO$_2$ multilayers, $k_{d,\text{Ag}}$ is 1.4 for the 30 nm-thick constituent layers, and 0.6 for 12 nm-thick layers. The phase shift in the thick-layered case is nearly a quarter turn, meaning significant interference effects that could hamper the coupling of metal surface plasmon resonances. The reason why the 12 nm-thick layers in AZO-ZnO remains true to EMT is because $k_{d,\text{ZnO}}$ is relatively small about 0.7. For 500 nm-thick layers of indium arsenide, $k_{d,\text{InAs}} = 1.04$. Therefore, what these may suggest is for the inequality to be modified to $k_d < k_{\text{z,EMT}} < \pi / 4$ or 0.8 to have reasonable agreement between ESLs determined by EMT and TMM. To gauge the phase shift within the multilayer slab determined by EMT, this inequality condition is reformulated to

$$k_{z,\text{EMT}}d_{\text{m}} << \pi$$

where $k_{z,\text{EMT}} = \sqrt{k_{d}^2 - k_{\text{z,EMT}}^2}$ employs the anisotropic permittivity of the multilayered medium. In the Ag-TiO$_2$ multilayers with 30 nm-thick layers, the left hand side of Eq. (5) is 0.4. For ZnO multilayers, this is 0.25, and for doped InAs multilayers it is 0.7. Therefore, the phase shift in both the constituent dielectric layers and the multilayer slab must be smaller than $\pi / 4$. The validity of EMT relies on the dielectric layer thickness, dielectric material permittivity, and the thickness of the overall multilayer film. It should be noted that in the case of near-field radiative heat transfer with hyperbolic modes or surface waves, evanescent waves are important and $k_D$ is usually imaginary. Therefore, the criterion needs to be modified [14,30].

3.5. Effect of Bloch waves

The dissipation of Bloch waves by the square of electric fields are illustrated in Fig. 5. The squared electric field magnitudes are normalized to that of the incidence field. For the Ag-TiO$_2$ multilayers, shown in Fig. 5(a), the waves formulated by EMT are continuous and periodic. The true Bloch waves are determined by TMM, which show dual-frequency envelopes due to two wave translation operators [10,31,32]. This dual-mode sinusoid is clear for the case when $d_{\text{m}} = 12$ nm, but not in the case of thicker layers. No clear frequencies or periodic behaviors are observed when the layer thickness becomes very large, meaning Bloch waves are no longer conserved. In Fig. 5(b), the Type II regime inhibits formation of Bloch waves due to $k_d$ purely containing an imaginary term.

In the AZO-ZnO multilayers, as shown in Fig. 5(c), despite having thick layers, the periodic but discontinuous Bloch waves are conserved throughout the thin film. The discontinuities in the Bloch waves are due to the lossless dielectric function used in modeling the dielectric layers. The discontinuous waves are also observed in Fig. 5(e), but for the 500 nm-thick layers, the periodicity and consistency are not as clear. Toward the bottom of the thin film, the Bloch waves begin to break down, leading to mismatching with the waves determined by EMT. The breakdown of Bloch waves may be understood by the attenuation of radiation over the course of the thin film. The two semiconductor multilayers share similar exponentially decaying fields in the Type II regime. Fig. 5(d) and (f) show the AZO-ZnO and indium arsenide multilayers, respectively. Agreeing with Fig. 4, the latter semiconductor multilayer is expected to decay faster and thus contributes less to the outgoing electric fields.
3.6. Penetration depth

The illustrate at which layer or period the breakdown of Bloch waves occurs, the penetration depths are studied, as shown in Fig. 6. Generally, the penetration depth should overestimate that by EMT, because the dielectric layers at least in the first few periods are nearly lossless. For Ag-TiO₂ multilayers shown in Fig. 6(a), the penetration depth profile for 30 nm-thick layers demonstrates a step-like reduction with incidence angle. The steps are smoothened because the dielectric is modeled as having a small loss. The Ag-TiO₂ multilayer films are 480 nm thick, which is below the penetration depth determined by either EMT or TMM. The consequence of large penetration depths relative to the multilayer slab thickness is the increased effect from multiple reflections. Given large phase shifts, the interference within a single homogeneous medium cannot be similar to that in the intricate multiply-reflected stratified layered system. EMT cannot account for the internal reflections, and therefore generally not suitable for determining penetration depths of multilayers. In the case of a Type II wavelength, the penetration depth is very small. In the case of the Ag-TiO₂ multilayer in Fig. 6(b), the penetration depth is no more than the thickness of the topmost layer, for all sensitivity

Fig. 5. Squared electric field magnitudes normalized to the incidence surface. The wavelengths and thin film thicknesses correspond to those in Fig. 3.
cases. There is hardly any variation with incidence angle since the medium appears as a dielectric thin film backed by a reflecting metal. EMT over-predicts the penetration depth because the imaginary components of TiO₂ and silver are small past wavelengths exceeding 400 nm [33].

In the semiconductor-dielectric multilayers, the penetration depths are comparable to the overall thickness of the thin film. The interference effect is evident in the step-down pattern with respect to the incidence angle. Fig. 6(c) and (e) show penetration depths in Type I wavelengths for Ag-TiO₂ and indium arsenide multilayers, respectively. The vertical drops in penetration depth correspond directly to the thickness of the constituent layers. Near normal incidence, penetration depth favors thicker constituent layers because fewer field-cancelling internal reflections between interfaces are encountered. For the Type II regime of semiconductor multilayers, as shown in Fig. 6(d) and (f), the penetration depth is smaller than the slab but greater than the constituent thicknesses. Here, unlike the case presented in Fig. 6(b), EMT under-predicts because the attenuation in the doped layers is significant. Only when the penetration depth is more than two orders magnitude larger than the constituent layer size, can EMT be used with the condition that the penetration depth is

Fig. 6. Penetration depths varying with the incidence angle. The wavelengths and thin film thicknesses correspond to those in Fig. 3.
shallower than the multilayer slab thickness. The validity of EMT in constructing ESLs relies on not only on small phase shift between layers, but also thickness of the multilayer slab compared to the attenuation factor.

4. Conclusions

This work helps the understanding of the effect of material or layer properties on estimating the optical and radiative properties in negative-refraction metamaterials. The use of EMT for the metal-dielectric (Ag–TiO₂) multilayers for visible wavelengths may not be well justified due to the large deviation from predictions using TMM. A general restriction of layer thickness is provided, alongside the demands of the material permittivity. The product of the wavevector and thickness of constituent layers or entire slab must be smaller than a quarter wave turn (κ/4). Since the largest wavevector magnitude relies on the dielectric layer’s permittivity, it is hinted that evanescent wave coupling between the metal layers are attenuated by the lossy dielectric. This decay is evident in the Bloch wave plots, which show loss of periodicity in multilayers are attenuated by the lossy dielectric. This decay is evident in the Bloch wave plots, which show loss of periodicity in multilayers containing lossy dielectrics. While semiconductor-dielectric multilayers may offer practical advantages, not all cases can justify the use EMT in estimating radiative properties. Further refinement of semiconductor and dielectric permittivity tuning can lead to better future consumable optical devices.

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